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STRUCTURE OF H2-O2 FLAMES UNDER PRESSURE.(U)
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STRUCTURE OF H_2O_2 FLAMES UNDER PRESSURE

Final Report

by

I. Sorgato

May 1981

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EUROPEAN RESEARCH OFFICE

United States Army

London England

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Particularly described are : the reactor and subsidiary equipment, the equipment and measuring methods of temperature profiles, concentration of radical species, combustion velocity and finally a new hybrid-stochastic method of flame simulation.

Particular emphasis is given to how much has newly been done from an experimental and theoretical point of view and to the results obtained in a field "flame under pressure", heterofore studied little. Several sets of complete measurements have been taken at various pressures up to 4 atmospheres.

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STRUCTURAL OF FLAME $H_2 - O_2$ UNDER PRESSURE

FINAL REPORT

The contract between the ERO London and Padua University - Institute of Chemical Plants refers to "Studies of the structure and composition of a flat hydrogen-oxygen flame under pressure by means of thermocouples and spectroscopic methods. The experimental results will be mathematically correlated".

ABSTRACT

The report illustrates the work carried out over 5 years, summarizing and correlating that which has already been set out in eight technical reports on the state of progress of the research with the addition of how much has been done since.

Particularly described are: the reactor and subsidiary equipment, the equipment and measuring methods of temperature profiles, concentration of radical species, combustion velocity and finally a new hybrid-stochastic method of flame simulation.

Particular emphasis is given to how much has newly been done from an experimental and theoretical point of view and to the results obtained in a field "flame under pressure", heretofore studied little, and in which the results given in literature are often contradictory.

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2. - MATHEMATICAL TREATMENT.
3. - RESULTS.

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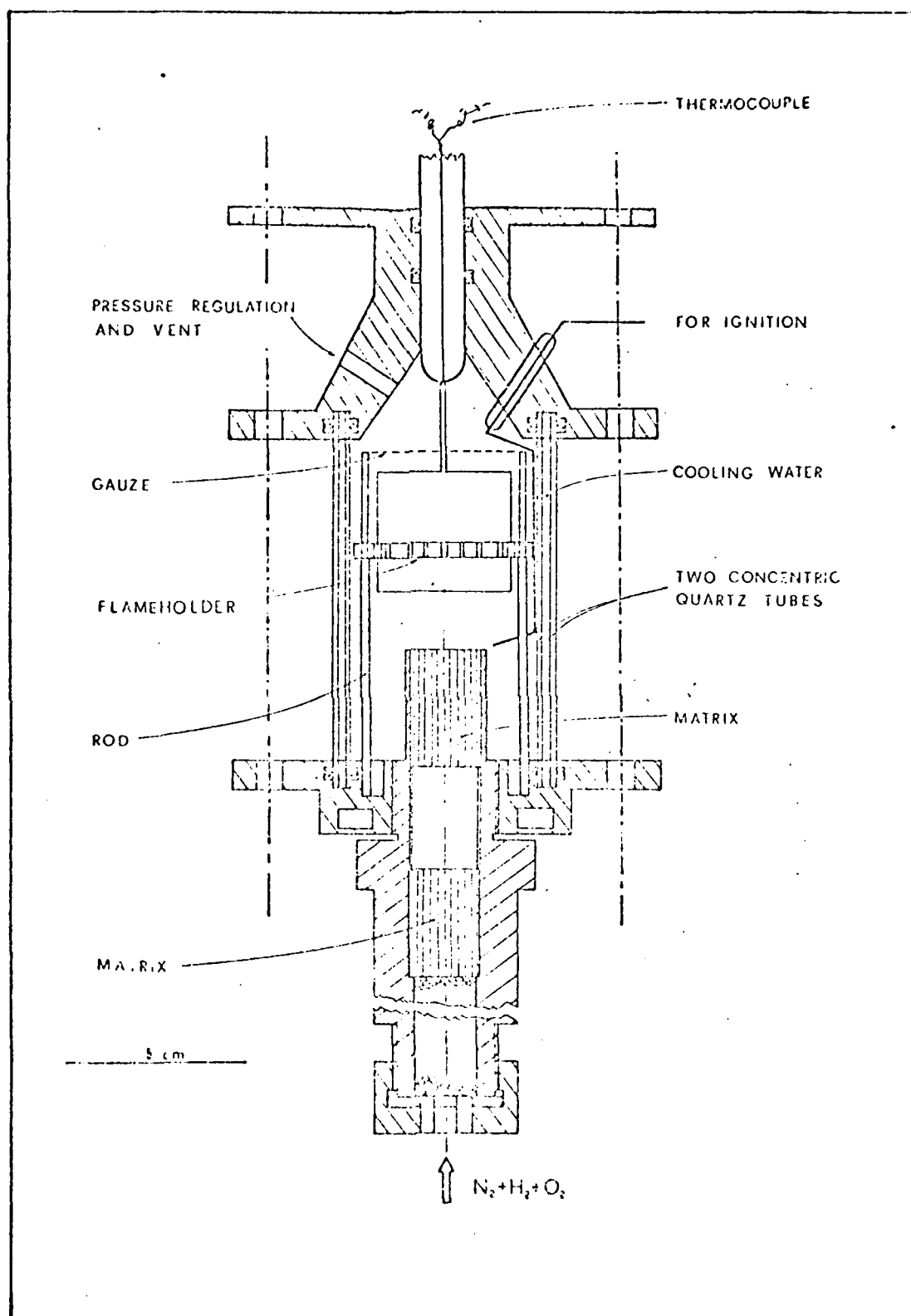


Fig. 1 - Flame transport reactor.

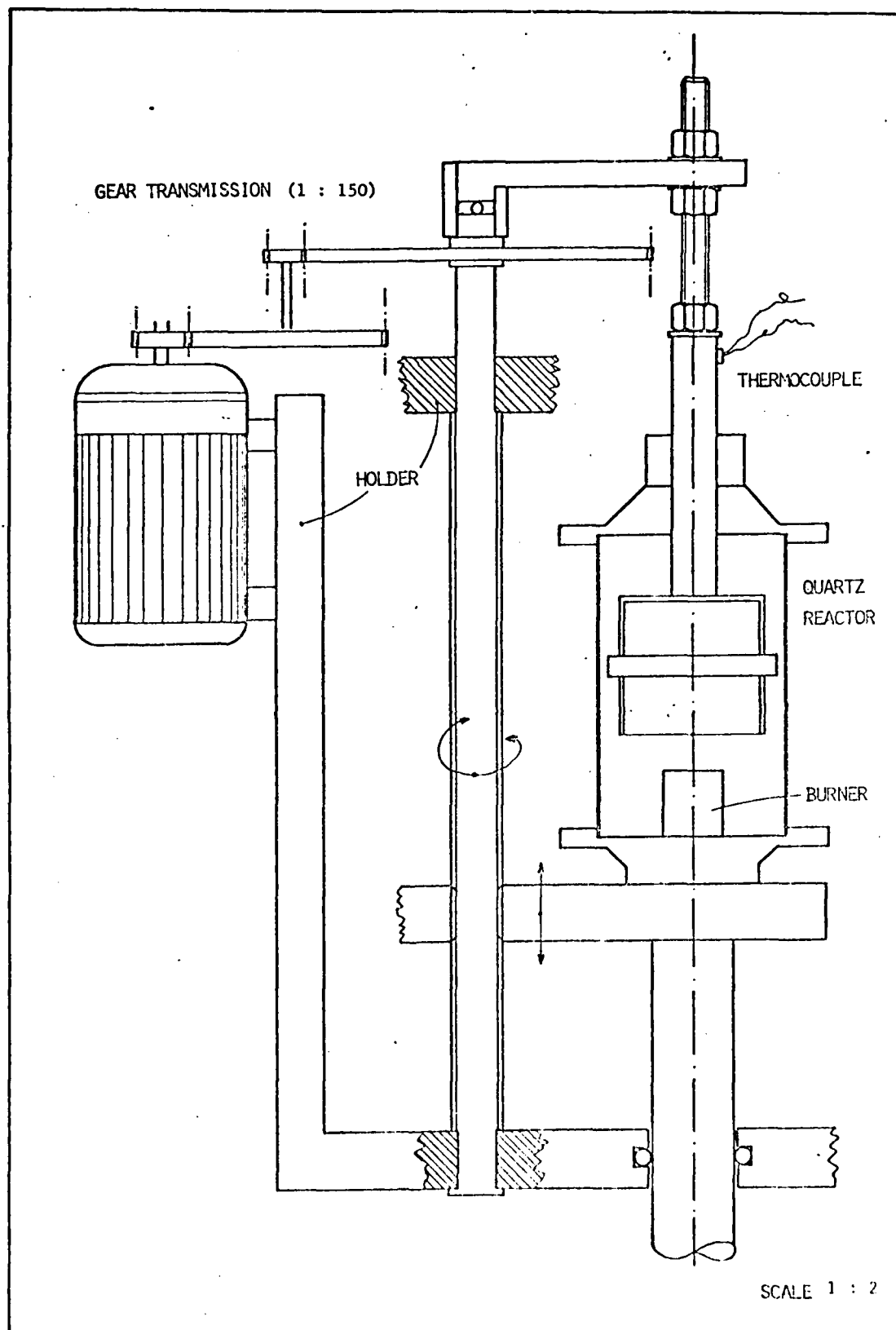


Fig. 2 - Automatic movement of the burner for the sealing of the profile.

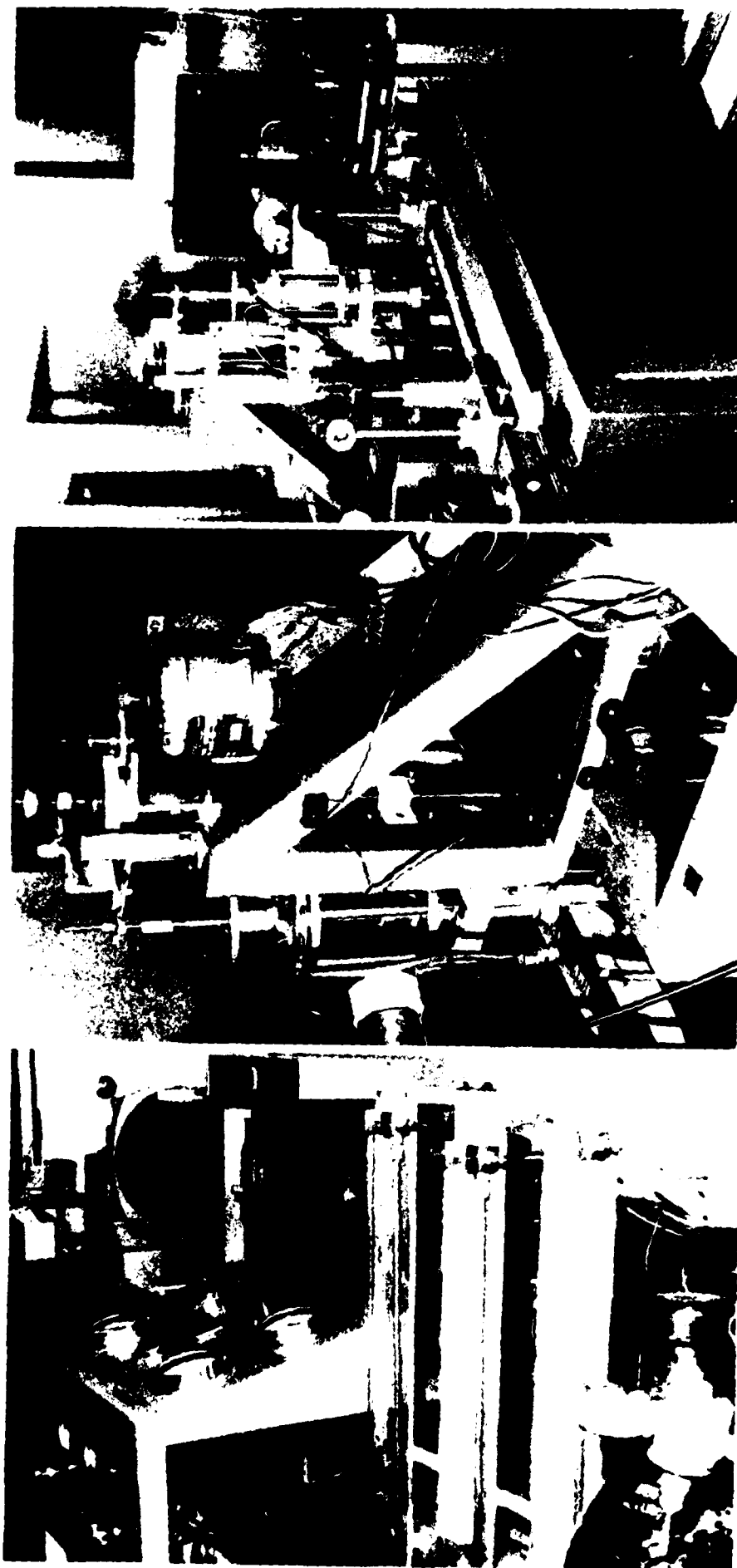


Fig. 2 - Details of the experimental set-up

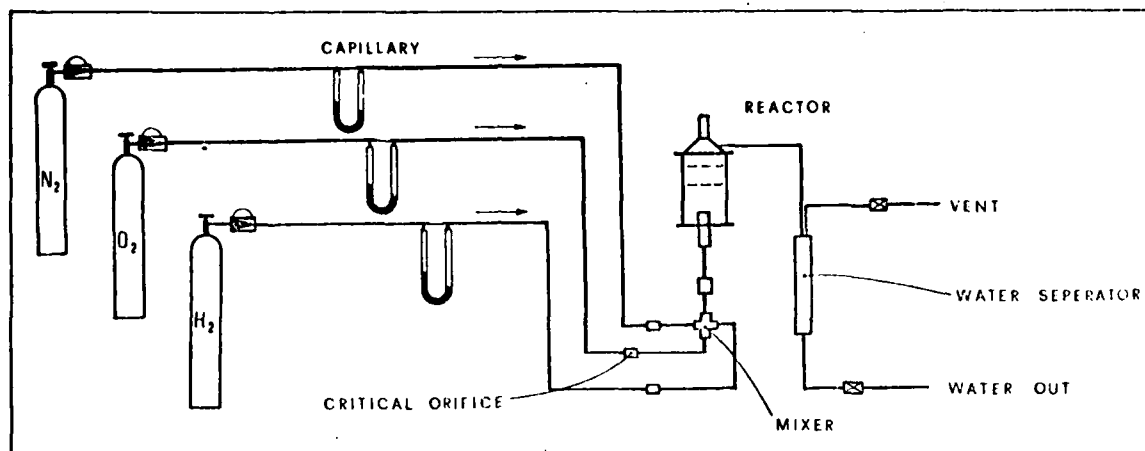


Fig. 4 - Gas flow system.

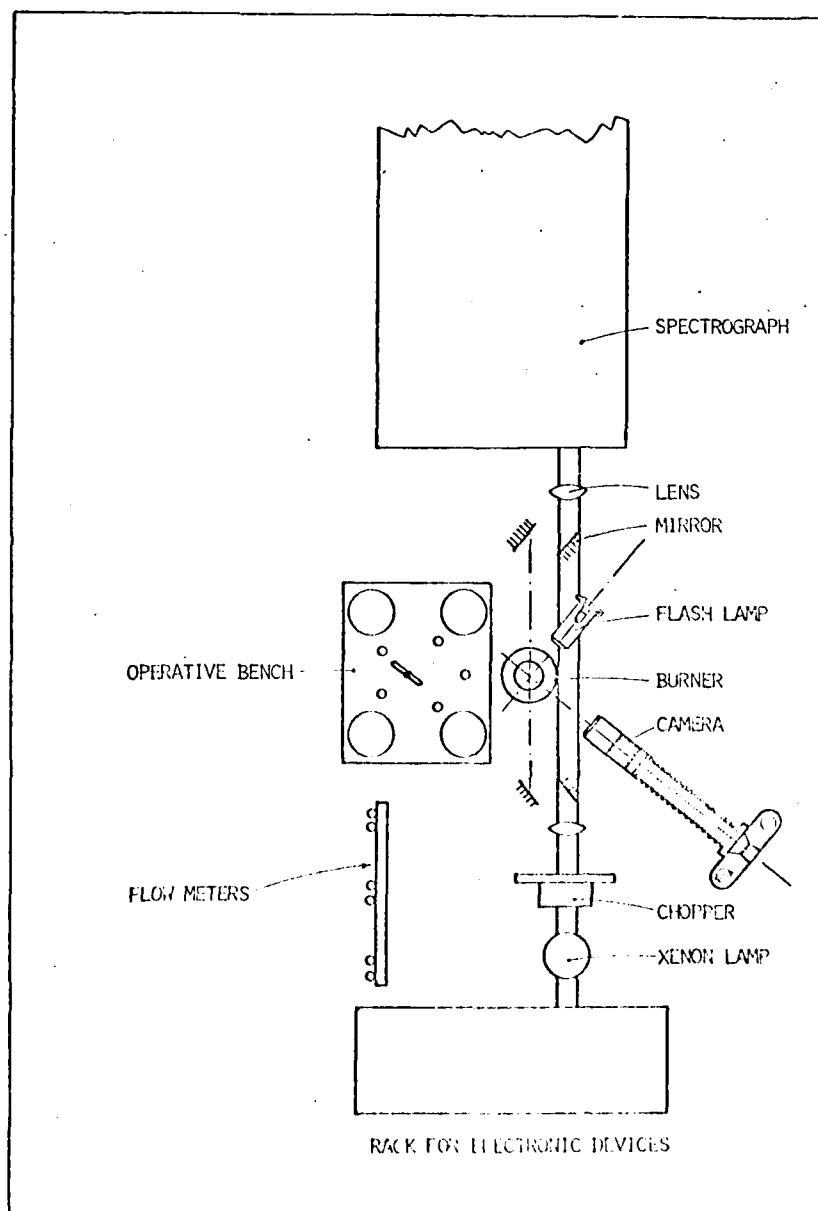


Fig. 5 - Laser Spectrograph Setup

1. - EXPERIMENTAL.

a) Reactor and subsidiary equipment.

The lay-out of the reactor built by us and used in the study of flames under pressure is given in Fig.1.

The characteristic parts are:

- the burner with corrugated matrix; which ensures a perfect mixing of the entering mixture and laminar plug flow in the reactant mixture.
- the combustion chamber, $\phi_{ins.} = 5.6$ cm, a double transparent quartz tube that allows working pressure up to $6-7 \cdot 10^5$ Pa and the carrying out of optical and spectroscopic measurements and observations of flame geometry and on the position of probes and thermocouples.
- the flameholder is a holed double disk in steel and pyrophyllite.

The combustion chamber is equipped with automatic scanning, Fig.2 which allows on one hand a high spatial resolution in the measurements by means of a thermocouple introduced in the flame as well as in the optical and spectroscopic ones, and on the other velocity in the experimental surveys thus ensuring constancy in the behaviour of the flame.

Fig. 3 shows the arrangement of the burner and instruments both for combustion speed, flame temperature and concentration of chemical species.

Fig. 4 and 5, in particular, show in detail the gas flow system and the optical instrumentation.

The various pieces of equipment are described in the paragraphs that concern the control of the progress of the reaction or measurement particulars.

b) Flow Velocity and Pressure Measurements.

Next to the reactor a critical orifice is set up which produces a pressure drop, so that the pressure in the combustion chamber is maintained at a constant value. Critical orifices are also placed in the feeding lines, these allow the flow velocity of the single components and the composition of the mixture to the burner to be measured. The pressure before and after is measured by transducers of the strain-gauge type (Philips Type PR 9800) set up in Wheatstone bridges on a thin sheet of steel.

The signals of the three feeding lines and that at the mixer outlet are measured on a magnetic "Tape" simultaneously with the temperature and

pressure values in the combustion chamber. The signals are also visualised on millimeters set up on the operative bench.

The transducers can visualise variations of few mm of Hg in normal working conditions up to $10 \cdot 10^5$ Pa.

c) Combustion Velocity.

It is known how this parameter is one of the most important in flame study. Data in the literature are scarce, in particular for combustion under pressure. Experiments, if conducted with accuracy, are the most trustworthy to obtain data on the combustion velocity of the mixture under examination.

The measurements are conducted visualising the flow by means of suspended particles of MgO. The stroboscopically illuminated particles are photographed every two seconds with 0.5 seconds exposure time. The camera is an Asahi-Pentax with a 135 mm F 2.5 telephoto lens. A double enlargement is obtained and a focal range from 1 to 2 mm. The range is focused on the luminous particles that are found on the axial diameter of the flame.

The velocity is determined by diagramming the height of the particles against time. The tangent to the curve at a point gives the combustion velocity at that point.

d) Temperature Profiles.

The temperature profile of a flame is that which analyses more completely its evolution, visualizing the preheating, reaction and post-reaction zones.

The temperature values along the flame are obtained through measurements by means of thermometric probes immersed in the flame and of special dimensions and shape, in order to disturb the gas flow as little as possible.

The probe used is a 25 microns diameter chromel-alumel thermocouple covered with a fused mixture of borax and alumina to eliminate the catalytic effects as much as possible. The very small diameter and the automatic scanning allow for good spatial resolution.

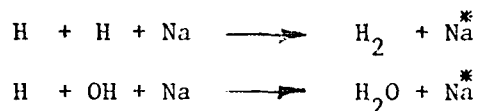
The instant position of the probe is recorded by the aid of a moving transducer which is capable of resolving the position with ± 10 microns accuracy. The effective measurement time is a few seconds, so as to ensure the constancy of the operative variables. Owing to the brief measurement time, it is not possible to use a normal pen recorder, whose constant is higher than that of the measurement, therefore a magnetic tape recorder (Analog 7 Philips) is used. The millivolts are transferred through a polynomial of

the 7th order which fits the data of the readings and the temperature obtained is then corrected for radiation losses of the thermocouple junction.

e) Concentration measurements of the OH and H species.

The concentration values of the radical species are an indispensable complement to the temperature measurement and a useful indication on flame behaviour, in particular in the reaction zone where the pressure effects can have a great effect on the reaction kinetics.

The relatively low flame temperatures $H_2-O_2-N_2$ do not allow the application of the emission spectroscopy except in the case of the fundamental lines of alkaline metals injected in the flame. This technique permits the determination of the H concentrations. The emission intensity of the sodium D lines is due only to the chemiluminescence according to the reactions:



since the thermal emission is irrelevant, because of the low temperatures of the flame.

On the other hand, absorption spectroscopy is used for OH concentration measurements. For such measurements a quartz xenon lamp is used as light source and a Philips XP 1003 photo-multiplier as transducer.

To raise the spatial resolution, cylindrical lenses focusing on the centre of the flat flame are used. The image on the entrance of the spectrometer a (Jarrel-Ash) is enlarged twice. The OH spectrum band (0,0) from 3060 to 3200 Å, is taken in the 2nd order to raise the resolving power of the spectrometer.

2. - MATHEMATICAL TREATMENT.

a) Heat Flow.

The volumetric chemical heat release rate results at any point

$$\dot{q}_r = \frac{d}{dz} \left(\dot{m} c_p T - \lambda \frac{dt}{dz} \right) \quad (1)$$

$\dot{m} = \rho_o v_o$ = mass flow rate of the mixture (ρ_o , v_o density and burning velocity at normal conditions)

c_p , λ = specific heat and local thermal conductivity.

Equations (1) describes the heat release rate as the gradient of energy flow due to the mass transfer (convection) and to the heat conduction. It does not make into account the heat losses which are due, in particular, to the radiation. The equation has been numerically calculated by Stirling's method.

The integrated heat rate througouht the flame interval

$$\dot{Q}_s = \int_{z=0}^{z=\infty} q_r dz \quad (2)$$

can be compared (apart from the small heat losses) with the heat calculated from the reaction enthalpy ΔH_r :

$$\dot{Q}_t = (V_o/V_{tot}) (\Delta H_r/M_{H_2}) \rho_o v_o \quad (3)$$

V_o , V_{tot} = volumetric flow rate of O_2 and total.

A comparison between the two heats allows good agreement with the temperature measurements.

b) Stochastic hybrid model of flame.

In the mathematical simulation of the burning process of a monodimensional flame stochastic solution has been adopted for the chemical process, which has, a more consistent physical basis than the commonly used deterministic. It adapts better to the use of the computer (avoiding "round-off" errors) and allows the use of calculating means to render the adopted model more exact and concise.

The stochastic solution presents a good elasticity so much so that it

has been possible to hybridise it with deterministic solutions for energy transfer and molecular diffusion.

Volume V, spatially homogeneous and in thermal equilibrium, contains a mixture of various chemical species which react among themselves in different ways. They are:

N_i = number of particles of the various chemical species S_i :

$$i = 1, 2, \dots, n.$$

R_r = chemical reactions, $r = 1, 2, \dots, m$.

The probability of the reaction R_r taking place is:

$$P_1(r) = Q_r / \sum_1^m Q_r \quad (4)$$

and the probable time is:

$$P_r(t) = \sum_1^m Q_r \exp - \left(\sum_1^m Q_r t \right) \quad (5)$$

where $Q_r = c_r f_r$, with f_r combinatory function of N_i and c_r , stochastic reaction constant = $k_r q! N^{g-1}$, $q = 1, 2, 3$ for different, two or three equal reacting molecules and g = reaction order.

In order to select the event and the probable time it is necessary to generate two casual numbers L_1 and L_2 included between 0 and 1 and to choose r so that:

$$\sum_1^{r-1} Q_r < L_1 \sum_1^m Q_r \leq \sum_1^r Q_r \quad (6)$$

and

$$t = (1 / \sum_1^m Q_r) \ln (1/L_2) \quad (7)$$

Once r is known the values of N_i are updated at time t according to the stoichiometry of reaction R_r .

Once the reaction enthalpies and the heat capacities of the various components are known, we have for every calculation cycle s , and for the adiabatic flame:

$$T_s = T_{s-1} - (\Delta H)_r / \sum_i (c_p N_i / N) \quad s = 0, 1, 2, \dots$$

and the average flow velocity will be:

$$\bar{v}_s = \bar{v}_{s-1} \frac{(NT)_s}{(NT)_0}$$

The conservation equation for the species i in the case of "premixed, stationary, laminar, ideally monodimensional flame" is:

$$\frac{d}{dz} \left[n_i (v + v_d) \right] = \frac{dn_i}{dz} \quad (8)$$

with z = axial coordinate, n = mole density number, v and v_d = mean velocity and diffusion.

Introducing the fractional mass flux we have:

$$\frac{\rho_o v_o}{M_i} \frac{d G_i}{V} = d n_i \quad (9)$$

with $\rho_o v_o$ = mass flux velocity

G_i = weight fraction of species i in the flux.

M = molecular weight.

Since the variation of n_i takes place in a stochastic way, per unitary increase, the displacement of one molecule will happen when:

$$\frac{\rho_o v_o}{M_i} \frac{G_i}{v} \begin{cases} > 1 & \text{negative diffusion} \\ < -1 & \text{positive diffusion} \end{cases} \quad (10)$$

For the type of flame adopted the energy conservation equation, integrated with the boundary conditions $z = 0$, $T = T_o$, $\partial T / \partial z = 0$ offers

$$\frac{\Delta T}{\Delta z} = \frac{v_o \rho_o \bar{c}_p (T - T_o)}{\bar{\lambda}} + \frac{Q_r}{\bar{\lambda}} \quad (11)$$

with \bar{c}_p and $\bar{\lambda}$ = mean specific heat and average conductivity of the mixture.

Equation (11) is called upon only for unitary events linked with the initial reactions between molecules (reactions 9 and 10 of Table 2). When these reactions which are the most probable in the high part of the flame are defined by the programme, their stochastic time determines the entity of heat transfer.

In this way both for molecular diffusion as well as heat transfer a system of hybridization is chosen adopting the deterministic equations only for unitary events linked to the stochastic approach.

Thus it is possible to calculate:

- the number of events;
- the temperature and concentration profiles;
- the stochastic time velocity and the distance.

3. - RESULTS.

Among the numerous runs described in the 8 reports, two have been chosen which characterize better the various theoretical and experimental aspects and on the other hand are sufficient in describing the behaviour of a flat flame under pressure.

The characteristics are given in Table 1.

TABLE 1

Chamber pressure	Ingress Composition			Flow Velocity	Q _t . eq(3)	Q _s . eq (2)
	H ₂	O ₂	N ₂			
Pa 10 ⁻⁵	%			cm sec ⁻¹	Cal (cm sec) ⁻¹	
1	16.15	5.40	78.45	9.05	2.80	2.55
3	12.40	5.15	82.55	12.85	11.30	10.51

Characteristics of flame at atmospheric pressure and under pressure.

As regards the heat release in general, a discrepancy of almost 10% is found between the values calculated by equation (3) and those deduced by the temperature profile equation (2). The agreement is good, taking into account the the heat due to radiation is not calculated.

The reaction probability, the temperature profiles and the composition of the two flames have been calculated with the help of a Laben 70 computer.

It is assumed $N_{tot} = 50.000$.

Of the 38 reactions that can happen in flames H₂-O₂-N₂ only 10 are necessary to obtain satisfactory results from the simulation without making the calculation too involved. The 10 reactions are given in Table 2 together with the kinetic constants. These last are suggested by BAULCH-DRYSDALE - HOME-LLOYD (²).

Reaction enthalpies, specific heats and thermal conductivities are those given in the table by PENNER (³) except that for HO₂ the values given in ref. (⁴) have been used.

TABLE 2

Reaction	k cin. (cm ³ mol ⁻¹ sec ⁻¹)	ΔT, K	ΔH 298 (kJ/mole)
1) O ₂ +H → O+OH	2.2·10 ¹⁴ exp(-8450/T)	700÷2200	70.672
2) H ₂ +O → H+OH	1.8·10 ¹⁰ T exp(-4480/T)	400÷2000	8.255
3) H ₂ +OH → E ₂ O+H	2.2·10 ¹³ exp(-2590/T)	300÷2500	-63.304
4) H+HO ₂ → OH+OH	2.5·10 ¹⁴ exp(950/T)	290÷800	-159.979
5) H ₂ O+H → H ₂ +OH	9.3·10 ¹³ exp(-10250/T)	300÷2500	63.304
6) H+H+N ₂ → H ₂ +N ₂	2.5·10 ¹⁸ /T	300÷5300	-435.973
7) O ₂ +H+N ₂ → HO ₂ +N ₂	1.5·10 ¹⁵ exp(500/T)	300÷2000	-197.096
8) H+HO ₂ → H ₂ O+O	5·10 ¹² T ^{0.5} exp(-2000/T)	300÷350	-231.538
9) H ₂ +O ₂ → OH+OH	8·10 ¹⁴ exp(-22500/T)	298÷2000	78.927
10) H ₂ +O ₂ → H+HO ₂	5.5·10 ¹³ exp(-29100/T)	298÷2000	238.906

The diffusion velocity, owing to the large excess of N₂ in the flame, is approximated by:

$$(v_d)_i = - \frac{N D_{i,N_2}}{N_j} \frac{d}{dz} \frac{N_i}{N} \quad (12)$$

The binary diffusion coefficient is evaluated from the well-known equation.

$$D_{i,N_2} = 2.628 \frac{T^{3/2} [(M_i - M_{N_2}) / (2 M_i M_{N_2})]^{1/2}}{\sigma_{ij}^2 P \Omega^* (T^*)} 10^{-3} \text{ cm}^2 \text{ sec}^{-1} \quad (13)$$

The values in the constants are given in ref. (5) while the corrections caused by the presence of polar species are valued according to (6). In the simulation programme the reduced collisional integral Ω^* (T*) is fitted with a 10th degree polynomial expression in (T)^{-1/2} from the data in (7); the polynomial coefficients are given in Table 3

TABLE 3

a = 0.440149	b = 1.056917	c = -0.036973
d = 0.818693	e = -1.416095	f = 2.153107
g = 6.386901	h = -2.910819	i = -7.933206
l = -3.845665	m = 9.390701	

The programmes are given in the appendix.

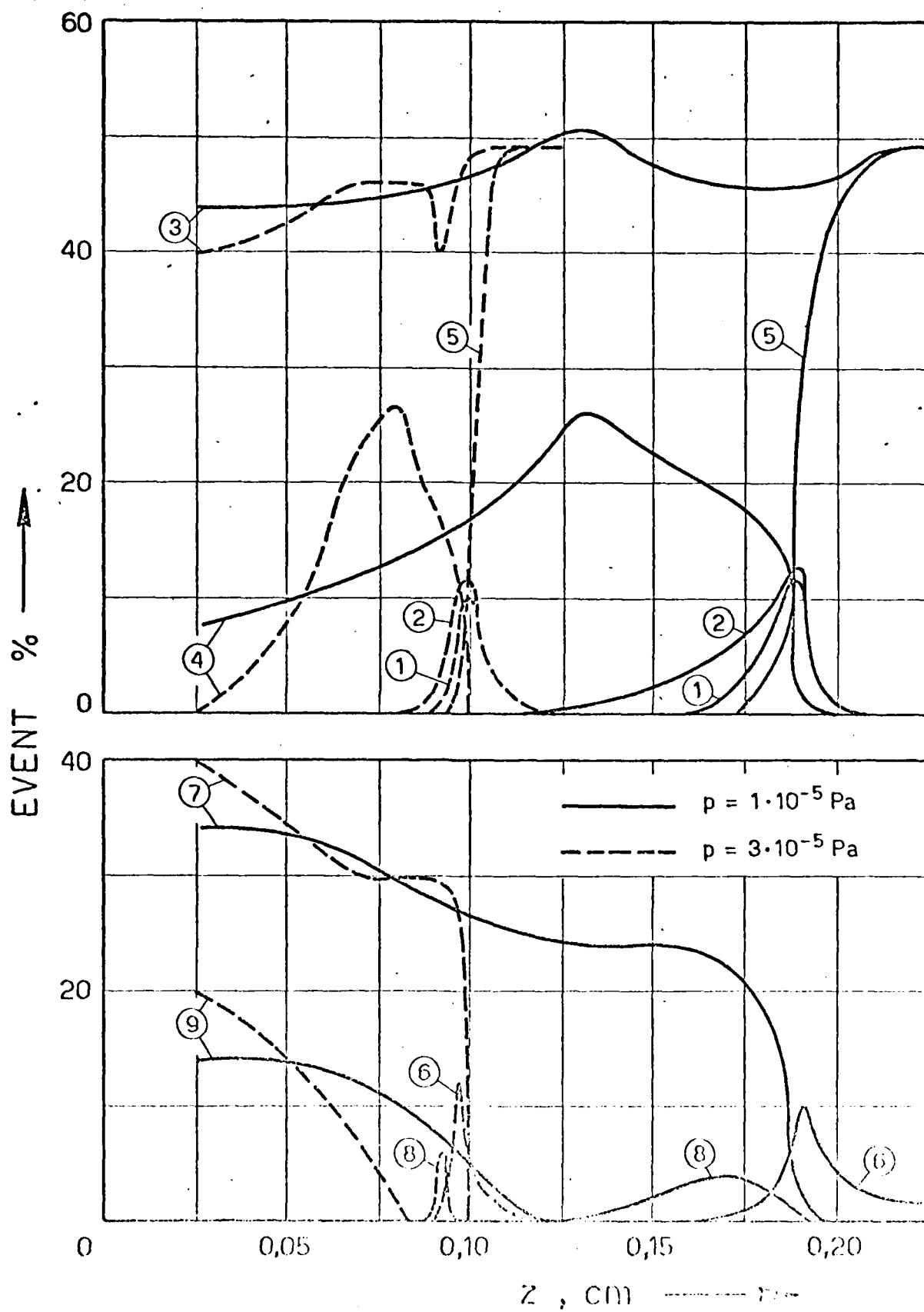


Fig. 6 - Events of the vacuum elementary reactions.

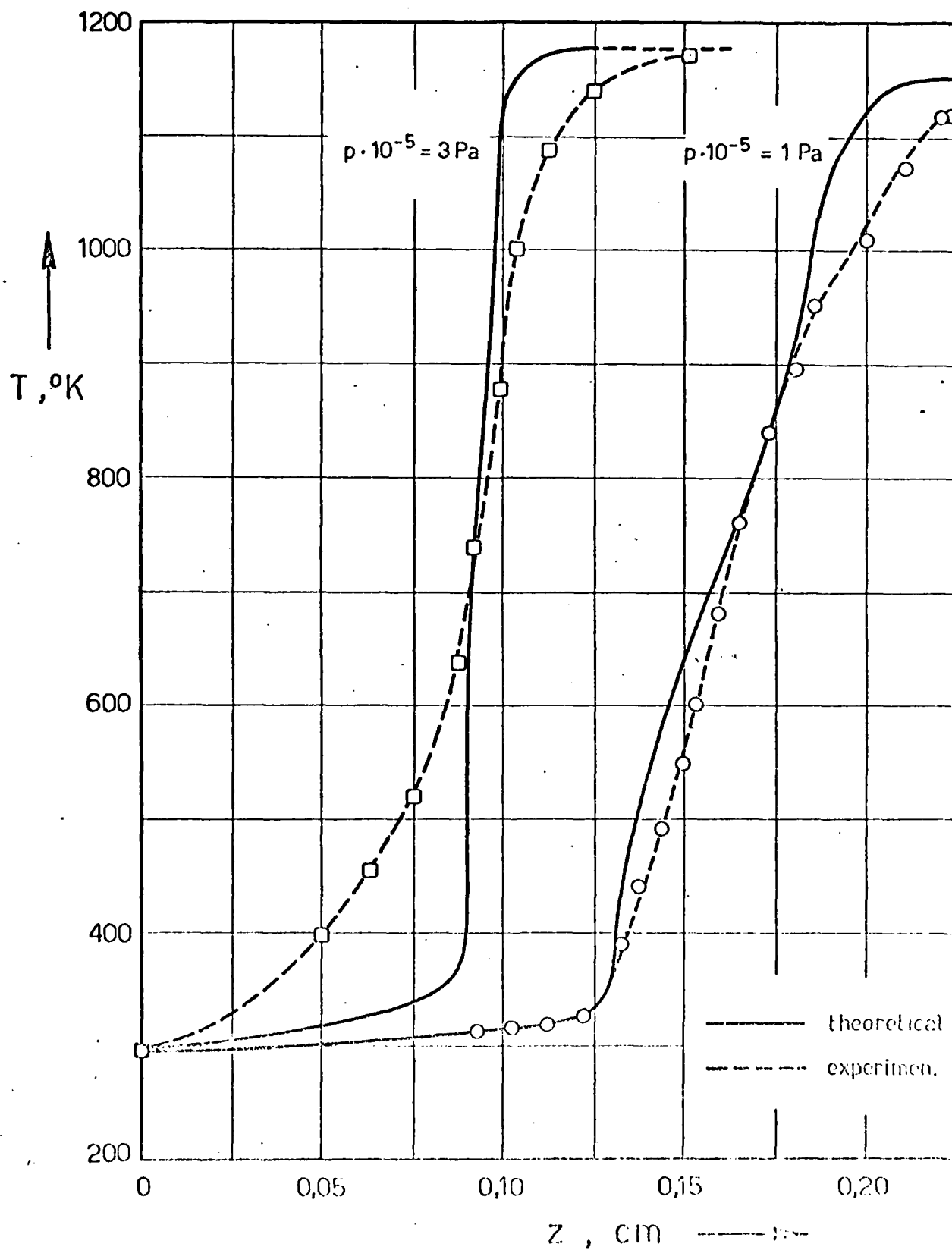


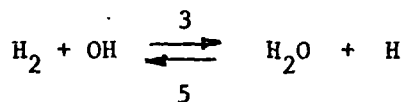
Fig. 7 - Temperature profiles.

a) Contribution of every single model reaction.

The calculation programme takes into account the stochastic path in the form of number of event of the various elementary reactions carried out by the same programme (Fig.6). Such data have allowed the programme optimization to reduce notably the troublesome kinetic model discarding the radical reactions which in our case were practically nil.

From the diagrams of Fig.6 it is possible to observe:

- the reactions between radicals tend to diminish at the approach of equilibrium. This can be represented by reactions (3) and (5).



- that reach in proximity of equilibrium, intervention values near to 100%.
- reaction (9) between H_2 and O_2 tends towards zero corresponding with the ignitions point of the flame.
- reaction (4) shows a maximum at intermediate temperatures, a zone in which the species H O_2 is stable, and which later decreases to zero at equilibrium.

b) Temperature Profiles.

The temperature profiles are given in Fig.7. A dotted line shows the experimental ones, a broken line indicates the calculated ones. The agreement can be regarded as satisfactory taking into account that:

- the experimental values suffer from approximate measurements and an imperfect flat shape of the flame.
- in the model the flame is assumed adiabatic and certain secondary reactions are disregarded.

From the two profiles of 1 and 3 atm it is possible to note, in agreement with the increase of the global reaction velocity with pressure: $\ln (r_1/r_2) = [(q/2) - 1] \ln (p_1/p_2)$ where q is the global reaction order = 2.081⁽⁸⁾, that equilibrium is reached more quickly as the pressure is increased and the reaction zone becomes thinner.

It is also noted (runs described in previous reports), that the effect of diffusion is more accentuated at low pressure than at high pressure, given that: the global reaction velocity is proportional to

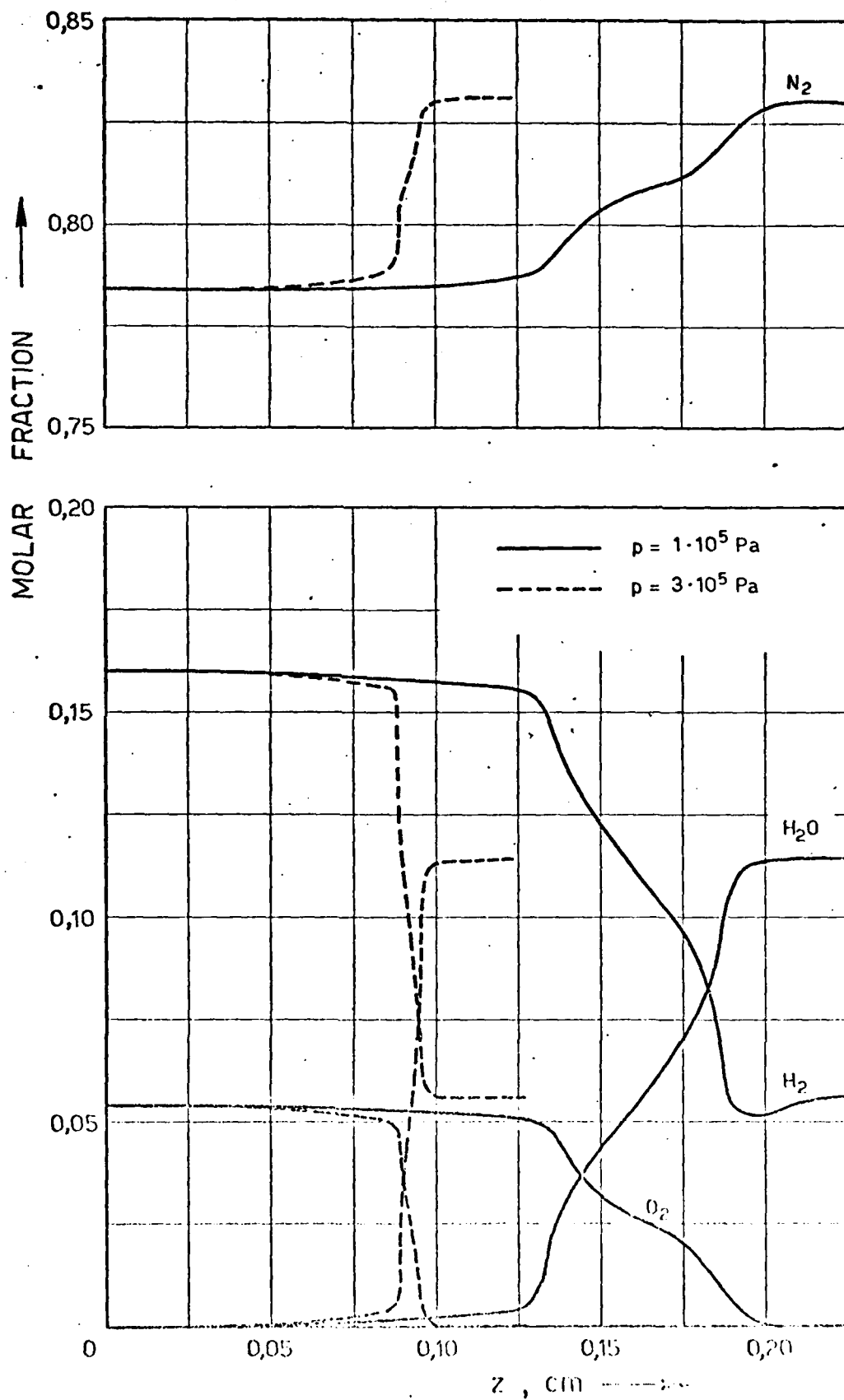


Fig. 6 - Concentration profiles (a).

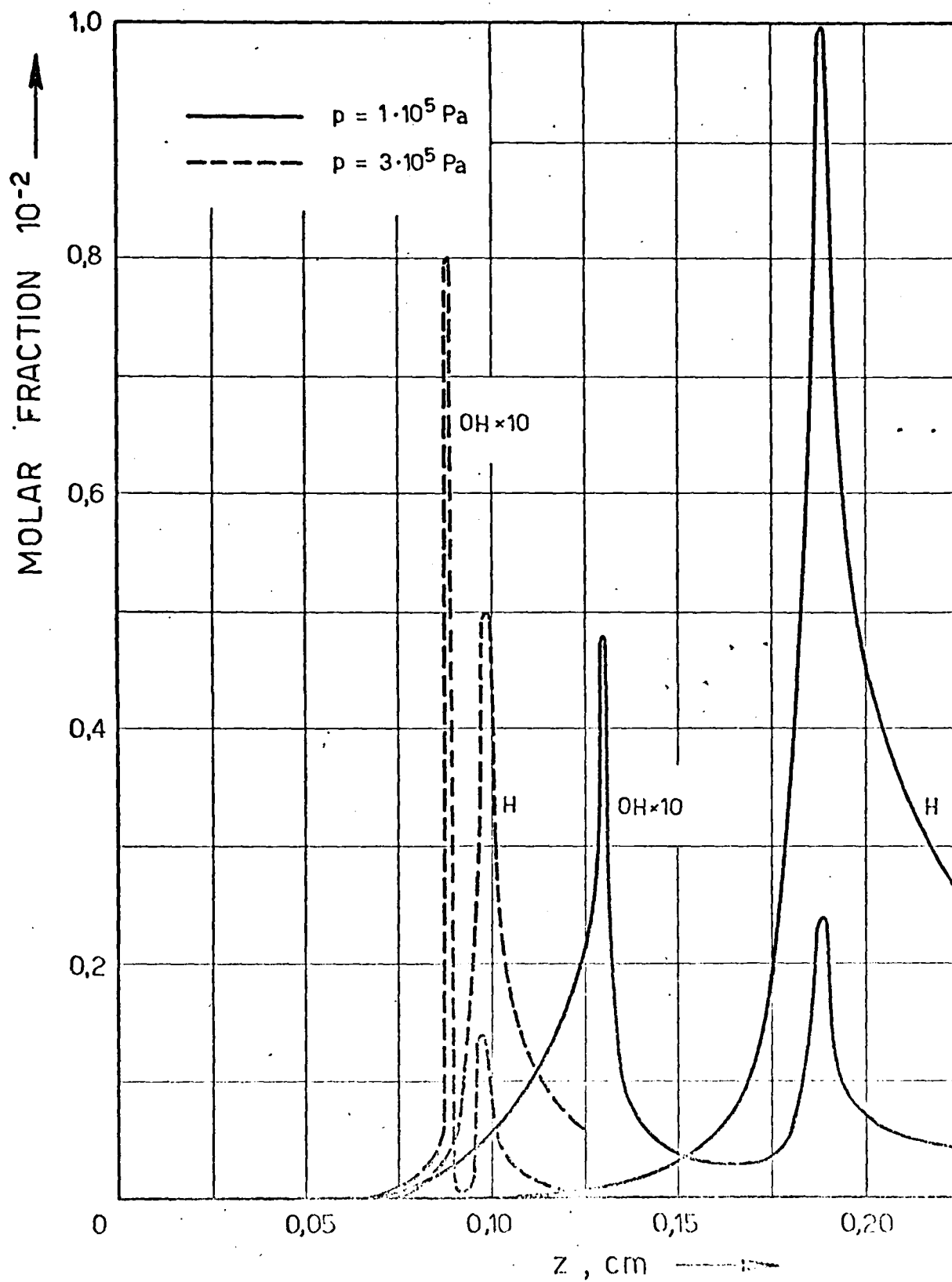


Fig. 9 - Concentration profiles (b).

$p^{(q/2)-1}$; the diffusion is inversely proportional to pressure, as it increases, the reactions gain in importance.

c) Concentration Profiles.

The facts previously described find verification also in the concentration profiles, Fig. 8-9. The increase of the global velocity leads to a quicker reaching of equilibrium conditions.

The two most important chain carriers, OH and H, show, as intermediate species, concentration maxima whose position depends on pressure. The relative curve at OH presents two of them, the first relative to reaction (4) maximum and the second together with that of H, due to reactions (1) and (2) maximum.

The presence of a minimum for OH had been noted in the experimental determination of such species, report 6.

CONCLUSIONS.

The theoretical-experimental study of flames $H_2-O_2-N_2$ under pressure has allowed us to identify the influence of the pressure parameter on fluid dynamics and on the progress of the system reactions in flow.

The small reaction zone of flame under pressure and the low temperature of flame due to the excess of N_2 are favourable conditions in the assumption of single dimensional flow and therefore ideal for the experimental check of the simulation model adopted.

This, a hybrid-stochastic type, offers better opportunities in respect of the deterministic normally used. In fact besides avoiding "round off errors", giving stable and accurate solutions, it is capable of indicating the most important physical phenomena and the most probable reaction mechanism for the various temperature and pressure conditions of the flame considered.

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